SUMMARY: SOME RESULTS FROM THE APIOS ATMOSPHERIC DEPOSITION MONITORING PROGRAM (1981-1988)

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ARB-101-92

Report Prepared By:

Air Resources Branch
Ontario Ministry of the Environment

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SUMMARY: SOME RESULTS FROM THE APIOS ATMOSPHERIC DEPOSITION MONITORING PROGRAM (1981-1988)

(1st Revised Edition)

ARB 101-92

Report Prepared By:

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and

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Air Resources Branch
Ontario Ministry of the Environment

92-04827/

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1. INTRODUCTION

This is an updated version, based on data collected from 1981 to 1988, of an earlier report that summarized some of the main results obtained from the Acidic Precipitation in Ontario Study's (APIOS) deposition monitoring program during the 1981-1984 period (Tang et al., 1986). The questions addressed include the current level of deposition of acidifying substances in Ontario, annual and seasonal variations of deposition levels, the relative importance of sulphur and nitrogen oxides as acid-forming substances, the relative importance of wet and dry deposition processes, and the relative contribution of sources in the United States and Ontario to provincial deposition levels. Some results are also given on the atmospheric deposition of the toxic metals lead and cadmium.

2. THE APIOS WET AND DRY DEPOSITION MONITORING NETWORK

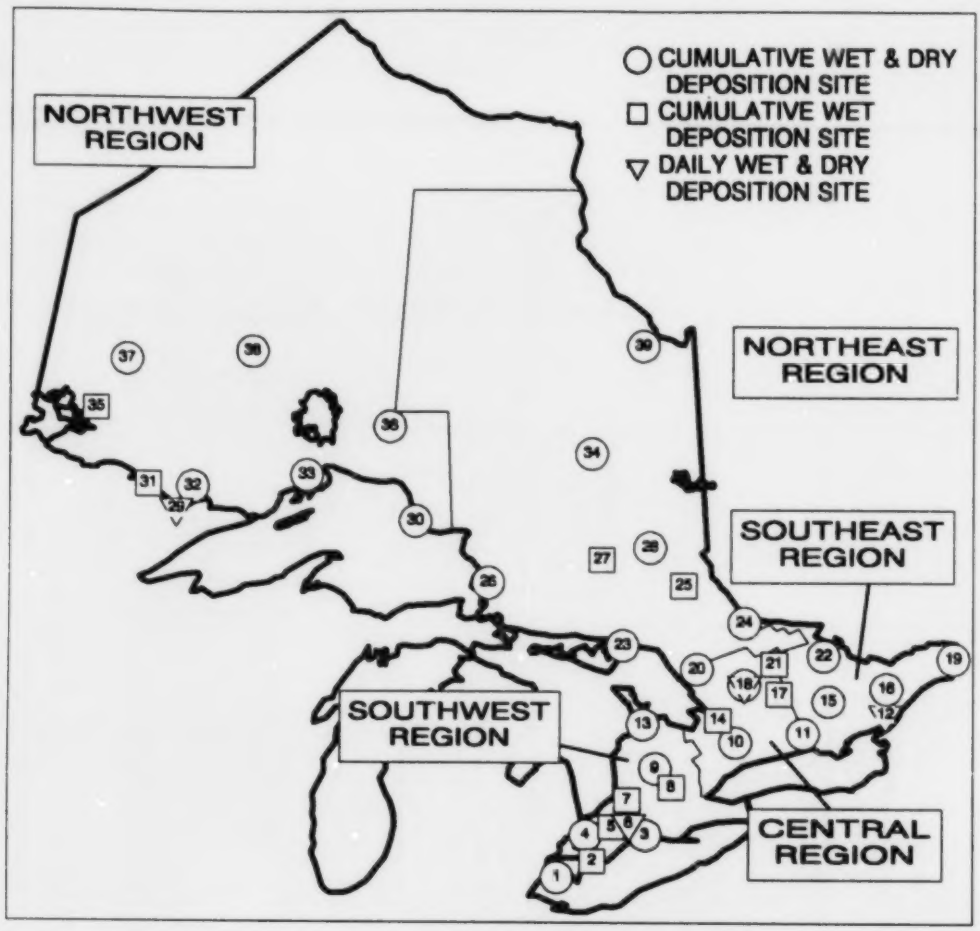
During the 1981-1988 period, atmospheric deposition was monitored at more than fifty rural locations in the Province of Ontario. Details of the deposition network have been given by Chan et al. (1985 a and 1985 b). Figure 1 shows the sites where sampling was carried out over a 28-day period and the sites where daily samples were collected. The 28-day, "cumulative" network is used to determine the long-term wet and dry deposition fields of acidity and related pollutants, such as sulphates and nitrates, as well as various toxic trace metals (such as cadmium and lead). The daily sampling network yields data on sulphur and nitrogen oxide concentrations which can be subjected to meteorological analysis. Air parcel trajectory analysis, for example, can be used to determine the contribution of sources in different compass directions to the atmospheric deposition at a receptor.

Precipitation is sampled using wet-only collectors, an example being shown in Figure 2. The collector vessel is normally covered, but when precipitation falls, a sensor (shown on the protruding arm) activates a motor, which causes the lid to be opened. When the precipitation stops, the lid is automatically closed. The sample is subsequently removed and sent to the laboratory for chemical analysis. The collector shown in Figure 2 is used in the cumulative network only. For the daily network, a slightly different design is used, but the principle of operation is the same.

Dry deposition of pollutants is not monitored directly in the APIOS networks - it is inferred from air concentrations. The dry deposition can be estimated for each pollutant if a quantity known as its "deposition velocity" is available. Then the dry deposition rate can be calculated by multiplying the air concentration and deposition velocity. This is possible for many of the pollutants monitored by the APIOS networks.

The equipment for monitoring air concentrations uses filter packs. Air is drawn by a vacuum pump through a series of filters (the first designed to remove particulates, and subsequent filters to trap nitric acid and sulphur dioxide respectively), which are later removed and subjected to chemical analysis at the Ministry's laboratory. Figure 3 shows the equipment used in the cumulative air monitoring network, where the air is continuously sampled over a 28-day period. The daily air monitoring stations have slightly different equipment, but the principle of operation is similar.

Fig. 1



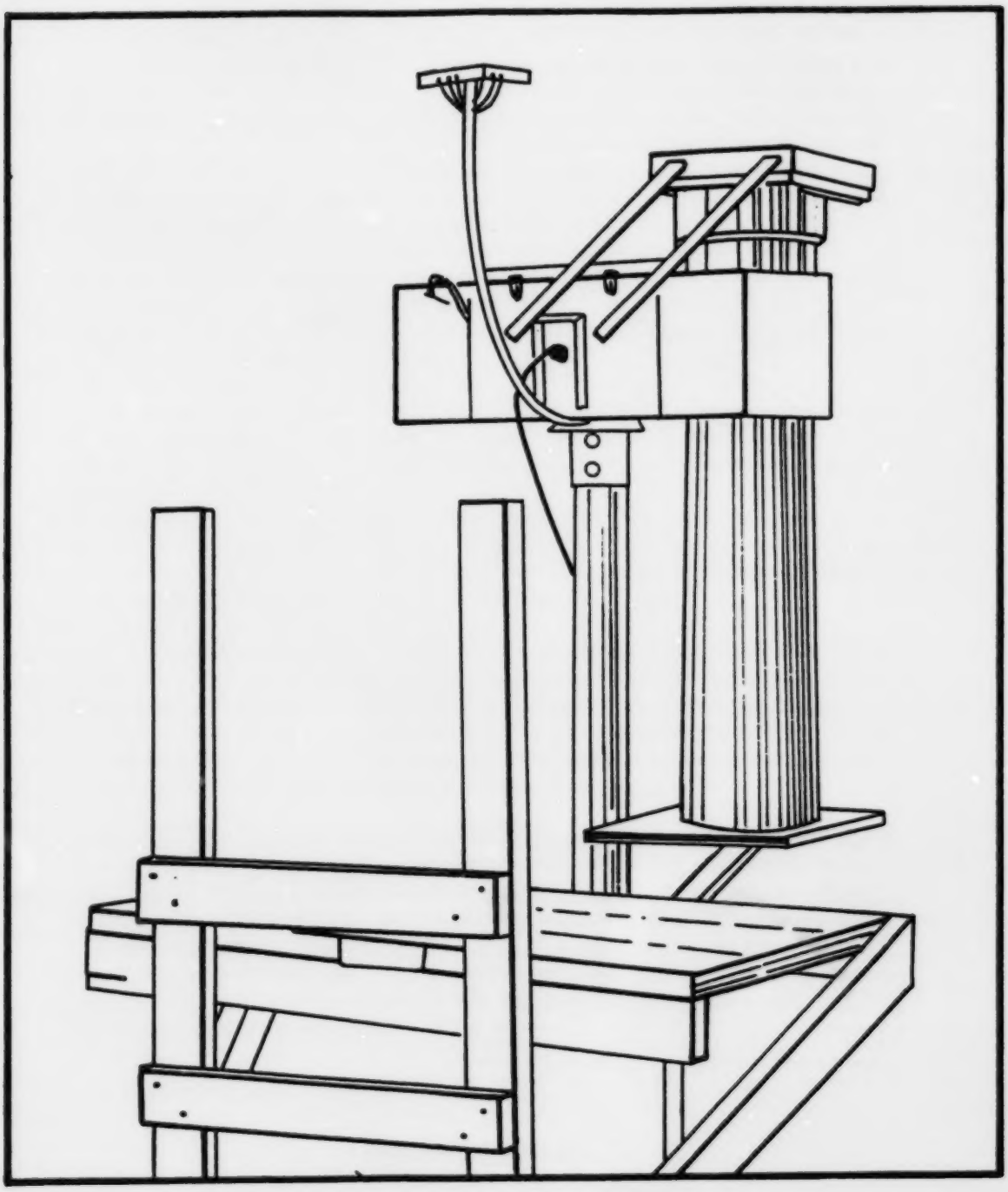
CUMULATIVE NETWORK SITES 1981-1988

1. COLCHESTER
2. MERLIN
3. PT. STANLEY
4. WILKESPORT
5. ALVINSTON
7. HURON PARK
8. WATERLOO
9. PALMERSTON
10. UXBRIDGE
11. CAMPBELLFORD
13. SHALLOW LAKE
14. COLDWATER
15. CLOYNE
    (replaced KALADAR, June 1983)
16. SMITH'S FALLS
17. WILBERFORCE
18. DORSET
19. DALHOUSIE MILLS
20. McKELLAR
21. WHITNEY
22. GOLDEN LAKE
23. KILLARNEY
24. MATTAWA
25. BEAR ISLAND
26. TURKEY LAKE
27. AZURE LAKE
    (replaced RAMSEY, June 1983)
28. GOWGANDA
30. OTTER ISLAND
    (operated only during 'summer' period)
31. LAC LA CROIX
32. QUETICO CENTRE
33. DORION
34. MOONBEAM
35. EXPERIMENTAL LAKES AREA
36. GERALDTON
    (replaced NAKINA, August 1983)
37. EAR FALLS
38. PICKLE LAKE
39. MOOSONEE
    (installed October 1985)

DAILY NETWORK SITES

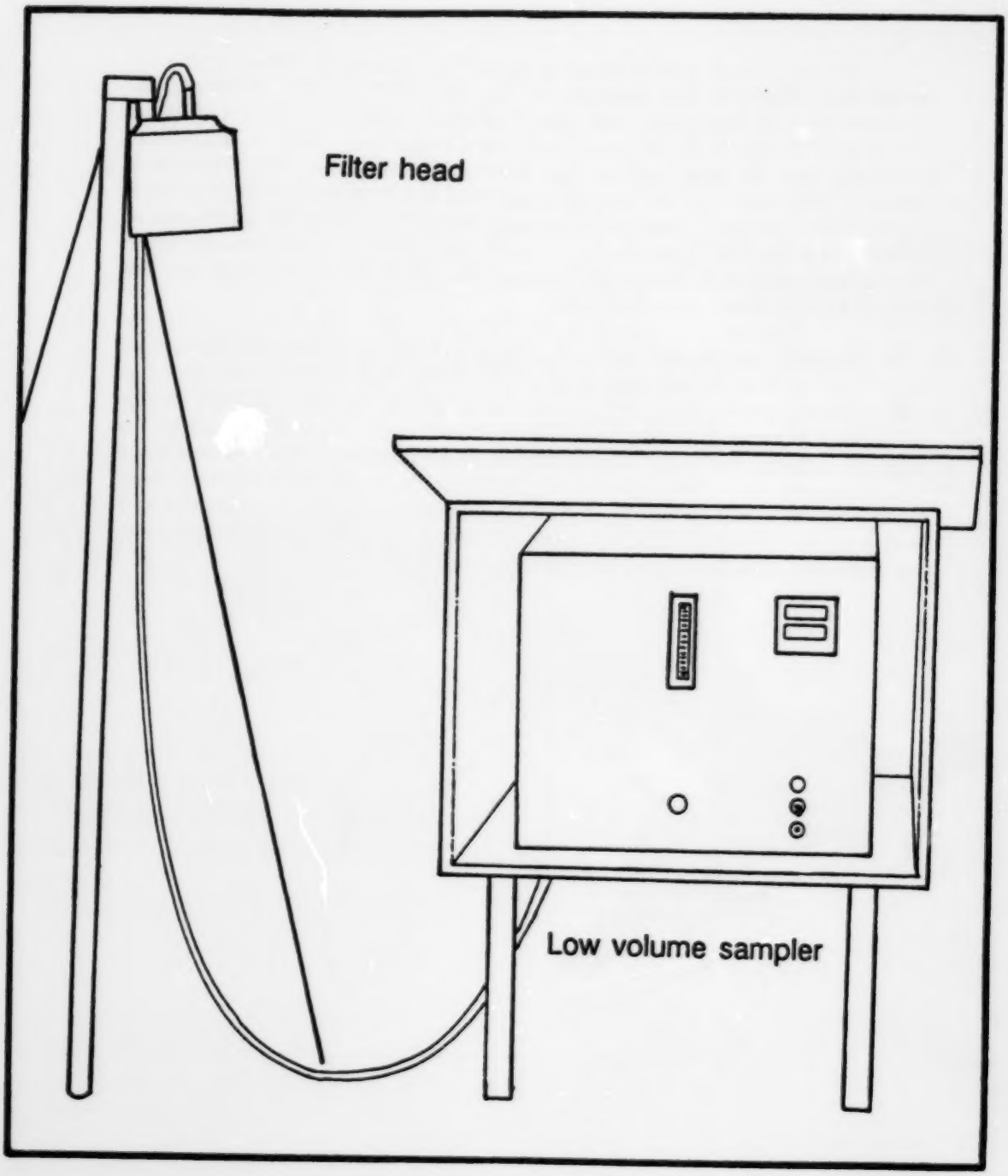
6. LONGWOODS
12. CHARLESTON LAKE
18. DORSET
29. FERNBERG

Fig. 2



THE APIOS PRECIPITATION COLLECTOR

Fig. 3



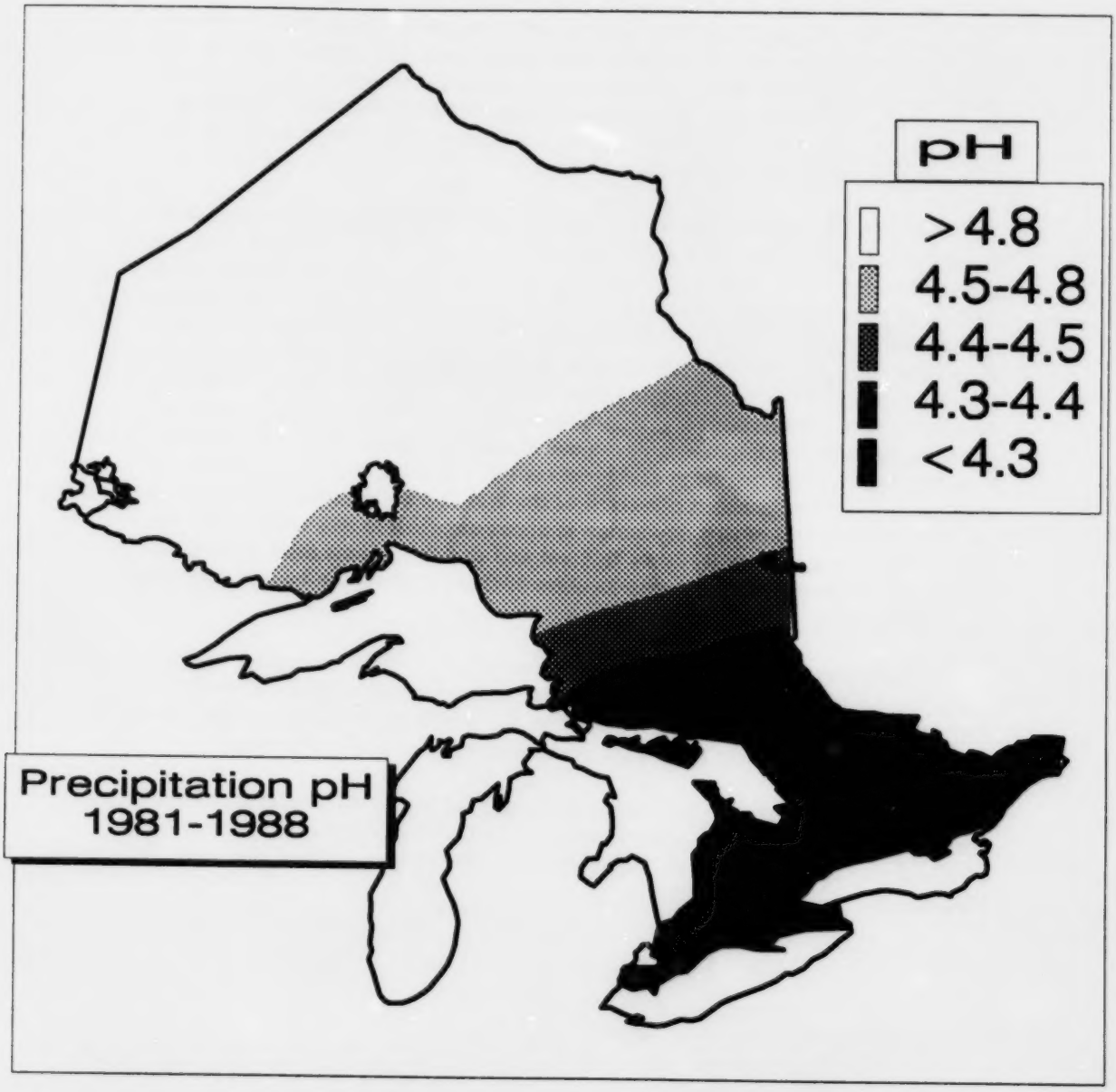
THE APIOS LOW-VOLUME AIR SAMPLER

3. PRECIPITATION ACIDITY

The acidity of precipitation is usually expressed in terms of the pH parameter, which is the negative of the logarithm of the hydrogen ion concentration. In very clean areas, the pH of precipitation is expected to be 5.6, due to dissolution of slightly acidic carbon dioxide gas, which is ubiquitous. However, even in areas remote from man-made influences, the presence of naturally emitted acidifying substances can lead to pH values as low as about 5. In polluted areas, precipitation acidity levels for individual rainfall events can be as much as a hundred times higher (i.e., pH values as low as 3.0), mainly due to the incorporation of sulphuric and nitric acids, which are formed from sulphur dioxide and nitrogen oxide emissions.

Figure 4 shows the long-term average pH values in Ontario, determined from data collected during eight years (1981-1988). Note that in most of central and southern Ontario, the average precipitation pH is presently less than 4.4 (i.e. the acidity is more than 16 times that of "clean" rain). The pH values in the Sudbury vicinity are lower than those of its surrounding area, (although the difference is small). This suggests that Sudbury has an influence on these data, though no effect is seen on the deposition of sulphate, which is presented later in this report. These are long-term average values. As was already pointed out, for individual events, the pH can fall considerably lower.

Fig. 4



4. SULPHATE WET AND DRY DEPOSITION

More than half of the acidity in precipitation comes from sulphuric acid. Actually, many scientists now feel that the environmental impact of acid rain can be inferred from the rate of sulphate wet deposition. An annual rate of wet deposition greater than 20 kg sulphate per hectare is thought to be harmful to sensitive aquatic ecosystems. As can be seen from Figure 5, this value is exceeded in all of central and southern Ontario. Figure 5 shows the annual sulphate wet deposition, determined from 1981-1988 data.

Acidifying sulphur species can also be delivered to the earth's surface by dry deposition processes - the direct absorption of sulphur dioxide gas from the atmosphere by water, soil and vegetation, and the impaction and gravitational settling of airborne sulphate particles. When these are taken up by the surface, the net result can be the same as when acid rain falls on the surface. Figure 6 shows the dry deposition rate of sulphur oxides, expressed in the same units as for wet deposition, i.e. as kilograms of sulphate per hectare per year. Note that dry deposition can deliver a significant additional loading of sulphate, over and above that due to precipitation (dry deposited sulphur dioxide is readily oxidized to sulphate). In southwestern and west central Ontario, the dry deposition of sulphur compounds is about one-third of the wet deposition rate. In the rest of the province the rate of dry deposition is about one-fifth to one-sixth of the rate of wet deposition. Figure 9 shows the wet and dry deposition rates of sulphate in different regions of Ontario (the location of these regions can be found on the map in Figure 1).

Note also that the atmospheric deposition of sulphur oxides is much greater in southern Ontario, near the emission source areas along the Great Lakes and in the heavily industrialized states to the south of the Great Lakes. This deposition pattern clearly illustrates the impact of man-made emissions.

(The figures shown for sulphate dry deposition in this report include the contribution of sulphur dioxide and sulphate particles. Sulphate deposition, both wet and dry, is expressed in kilograms of sulphate per hectare per year, kg SO_4/ha/yr.)

Fig. 5

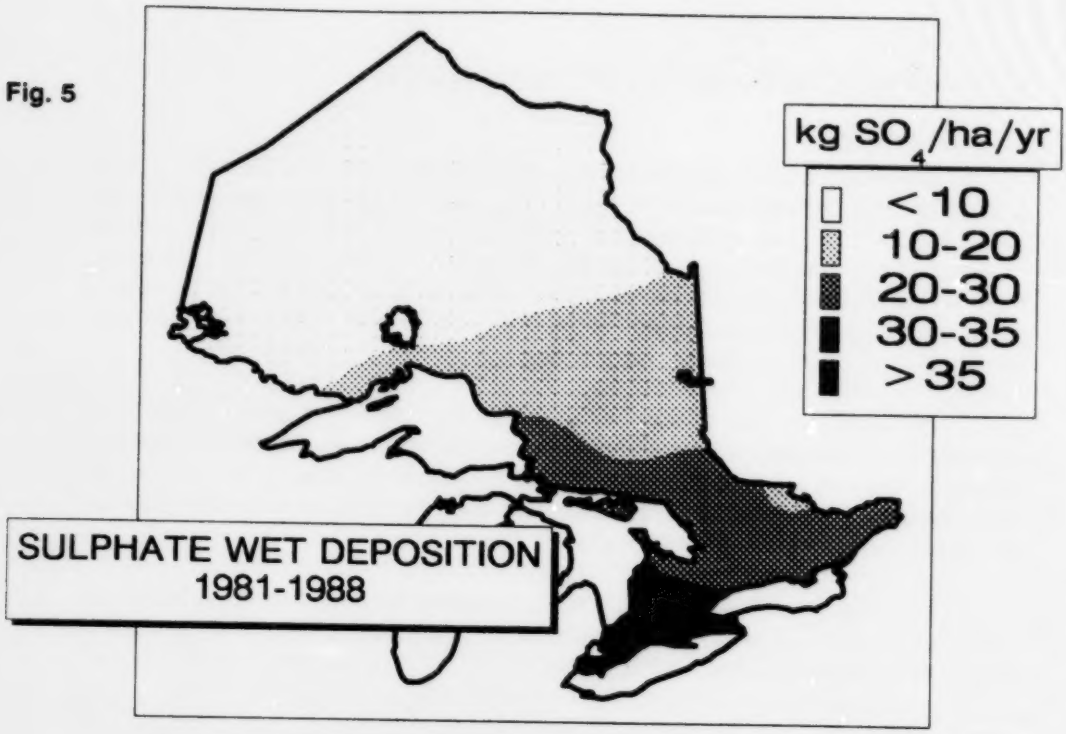
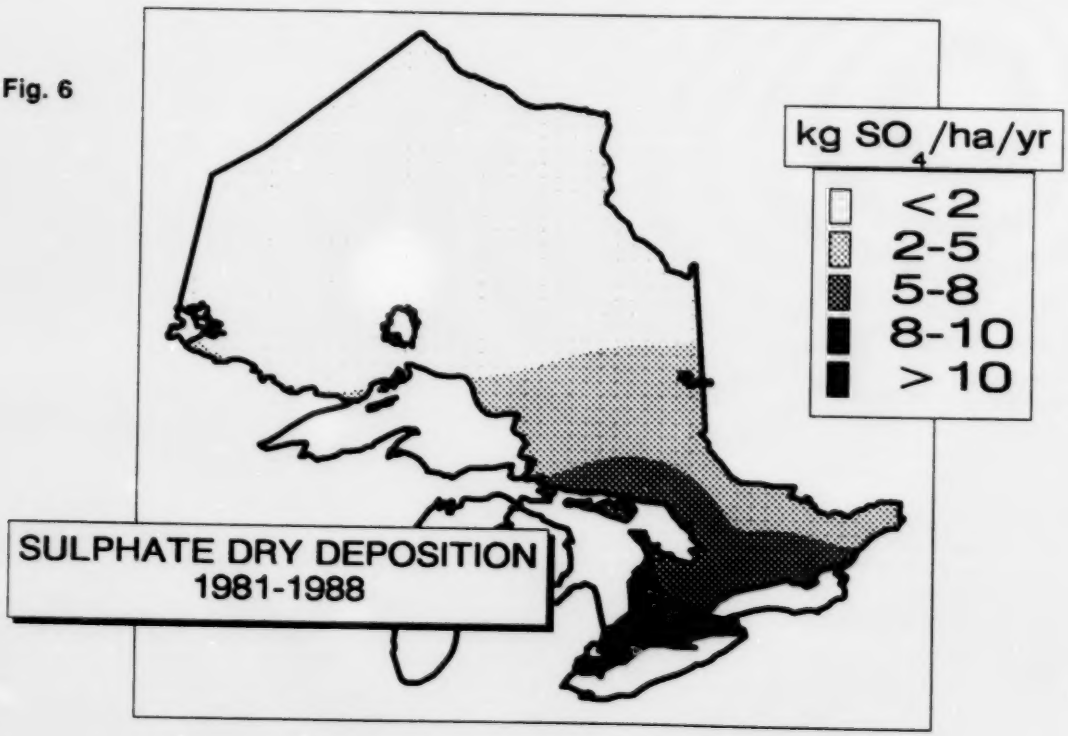

kg SO_4/ha/yr
< 10
10-20
20-30
30-35
> 35
SULPHATE WET DEPOSITION
1981-1988

Fig. 6


kg SO_4/ha/yr
< 2
2-5
5-8
8-10
> 10
SULPHATE DRY DEPOSITION
1981-1988

5. NITRATE WET AND DRY DEPOSITION

The other major contributor to precipitation acidity in Ontario is nitric acid. Figure 7 shows the annual nitrate wet deposition rate determined from 1981-1988 data. Figure 8 shows the estimated annual dry deposition rate for nitrates. A comparison of these figures shows that the wet and dry deposition rates are quite similar across the province. This is so because a large portion of the airborne nitrogen compounds is in the form of nitric acid, which is very readily absorbed by surfaces, hence giving a high dry deposition rate.

As with the case of sulphur compounds, there is a rapid drop-off of atmospheric deposition with distance from the emission areas in the northeastern United States and southern Ontario. Figure 10 illustrates (using 1981-1988 data) how the total annual nitrate deposition and the contribution from wet and dry processes, varies from region to region of the province.

(The figures shown for nitrate deposition in this report, include the contribution of nitric acid vapour and nitrate particulates. Nitrate deposition is expressed in units of kilograms of nitrogen per hectare per year, kg N/ha/yr.)

Fig. 7

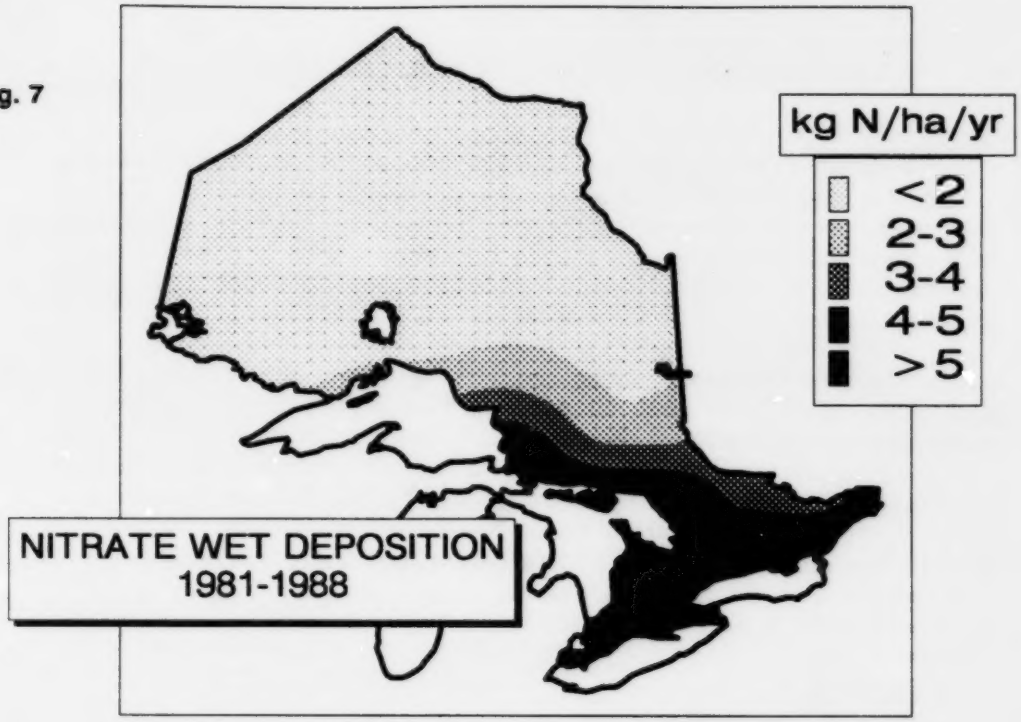
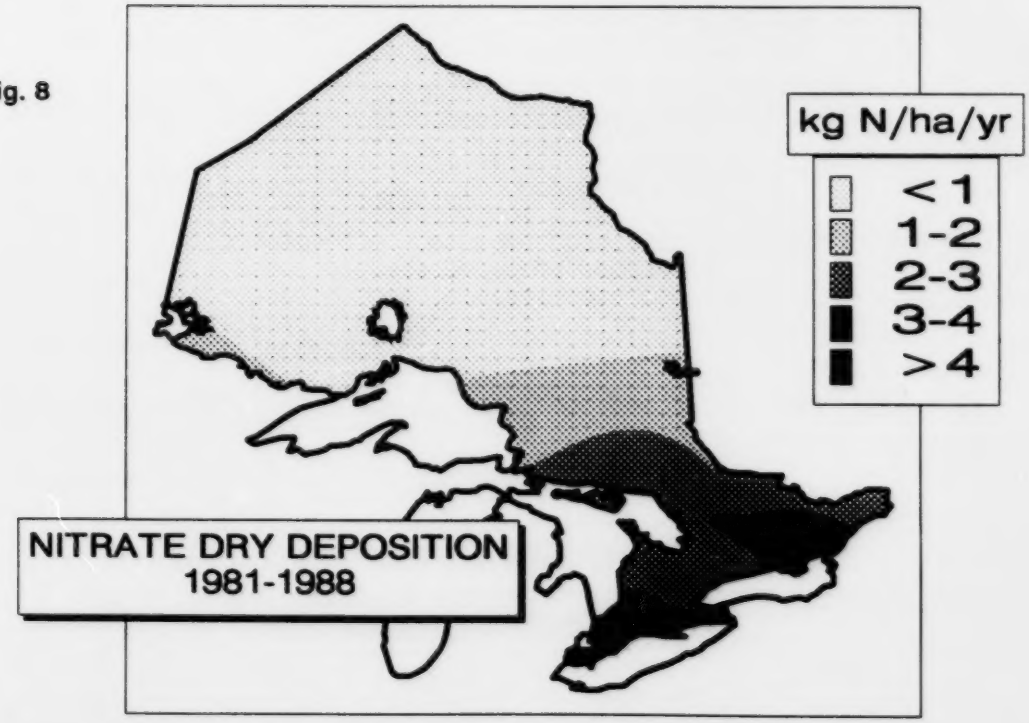

kg N/ha/yr
< 2
2-3
3-4
4-5
> 5
NITRATE WET DEPOSITION
1981-1988

Fig. 8


kg N/ha/yr
< 1
1-2
2-3
3-4
> 4
NITRATE DRY DEPOSITION
1981-1988

6. SULPHATES VERSUS NITRATES IN ACID DEPOSITION

Figures 5 to 10 may give a misleading impression regarding the relative contribution of nitrates and sulphates to acidic deposition, because of the different units used to express the results. To express the impact of sulphates and nitrates on a comparable basis, scientists convert the values shown in these figures to "equivalents". Each equivalent of sulphate has the potential of yielding the same amount of acidity as an equivalent of nitrate.

When the atmospheric deposition (wet plus dry) results are converted to equivalents of sulphate and nitrate, it is found that their potential for environmental acidification is comparable, i.e. almost as much acidity is due to nitrogen as sulphur oxides. This is illustrated in Figure 11 which shows the total deposition (wet plus dry) of sulphur and nitrogen oxides in different regions of the province, in terms of equivalents per hectare per year. Clearly, although the total atmospheric deposition of acidic-related compound drops sharply going from south to north, the contribution of sulphur and nitrogen compounds remains similar, about 54 to 61% of the acidifying potential coming from the sulphur oxides.

Fig. 9

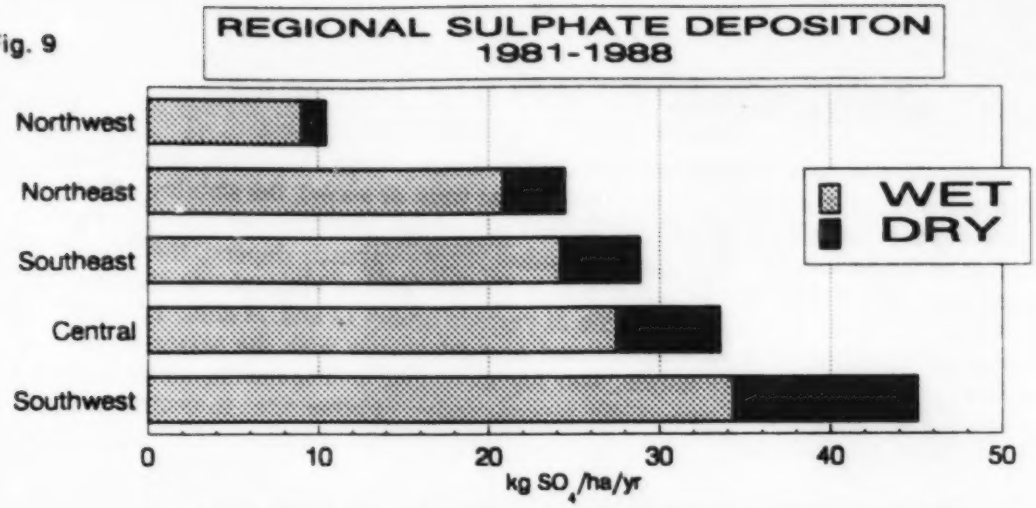


Fig. 10

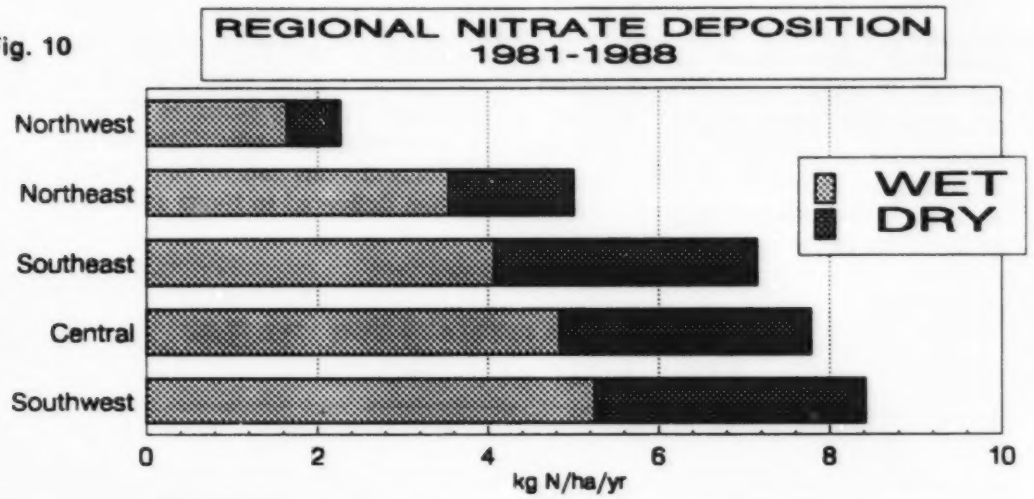
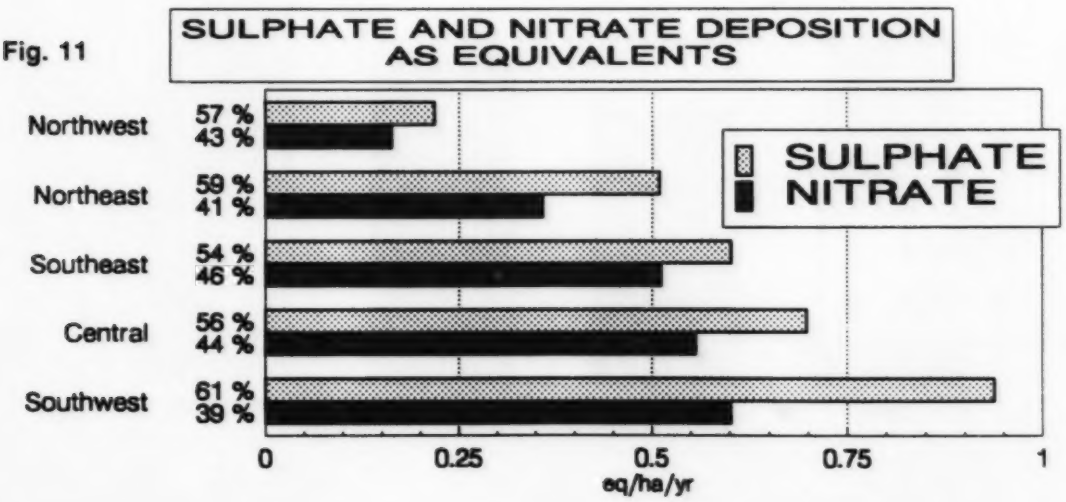


Fig. 11



7. SEASONAL TRENDS

Atmospheric deposition can change from season to season, since it is determined by the nature of the precipitation (rain or snow), the stability of the atmosphere (which can be quite different for winter and summer months), the state of the surface on which dry deposition processes partly depend, the rate of emission of sulphur and nitrogen oxides and the rate of atmospheric chemical reactions (which depend on the time of year). Figures 12 and 13 show how the atmospheric deposition of sulphur and nitrogen varies with the season at the APIOS Dorset monitoring station, near Algonquin Park. The results, taken from data over the 1981-1988 period, are representative of other sites in central and southern Ontario as well.

Note that for sulphates, there is a maximum in the atmospheric deposition during the summer months. For nitrates, there is a moderate increase of the total deposition in the winter. When expressed in terms of equivalent weights, the amount of acidity potentially delivered by nitrates during the winter season in about twice as great as that delivered by sulphates. This emphasizes that the importance of nitrogen controls should not be underestimated. The acidity accumulated in the snow pack during the winter months can lead to "acid shock", and result in damage to aquatic ecosystems, when delivered to rivers and lakes at the onset of the spring melt.

Fig. 12

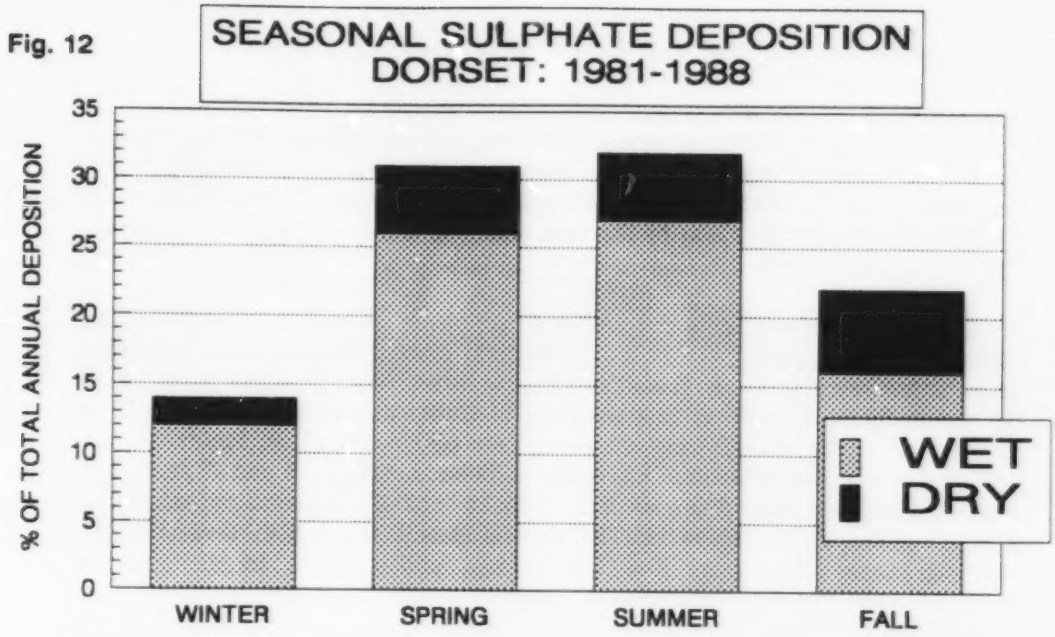
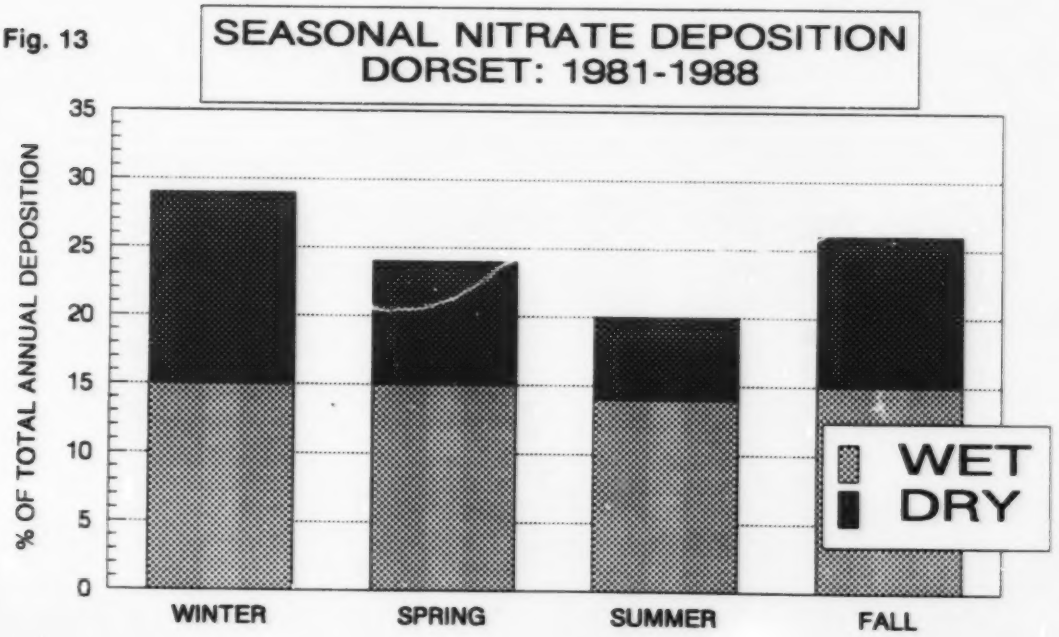

SEASONAL SULPHATE DEPOSITION
DORSET: 1981-1988
% OF TOTAL ANNUAL DEPOSITION
35
30
25
20
15
10
5
0
WINTER
SPRING
SUMMER
FALL
WET
DRY

Fig. 13


SEASONAL NITRATE DEPOSITION
DORSET: 1981-1988
% OF TOTAL ANNUAL DEPOSITION
35
30
25
20
15
10
5
0
WINTER
SPRING
SUMMER
FALL
WET
DRY

8. ANNUAL TRENDS

The amount of atmospheric deposition also varies from year to year due to, for example, changes in emision rates or meteorological variability. As everyone knows, there can be large changes in the amount of precipitation at a given location from one year to the next, and the wet deposition rate depends strongly on this factor.

Figure 14 shows how the annual atmospheric deposition of sulphates at Dorset has varied during the 1981-1988 period. Note that even when expressed on an annual average basis, there can be a considerable year-to-year variation in the atmospheric deposition (the seasonal or individual event variability is of course much greater). During the eight-year period shown in Figure 14, the annual sulphate wet deposition has fluctuated between about 25 and 42 kg/ha/yr with a fairly consistent decrease from 1981 to 1987. Corresponding values for nitrate are shown in Figure 15. There is no clear trend (up or down) in the nitrogen deposition rates. The year-to-year variability is largely due to meteorological factors, and makes it difficult to isolate the effects of emission controls (such as Ontario's "Countdown Acid Rain" program) on acid deposition. However, by accumulating a sufficiently long data record, and by applying special data analysis techniques, these effects can be determined. Our network is closely monitoring the results of Ontario's emissions control program, as well as the effects of acid gas abatement programs in the United States.

Fig. 14

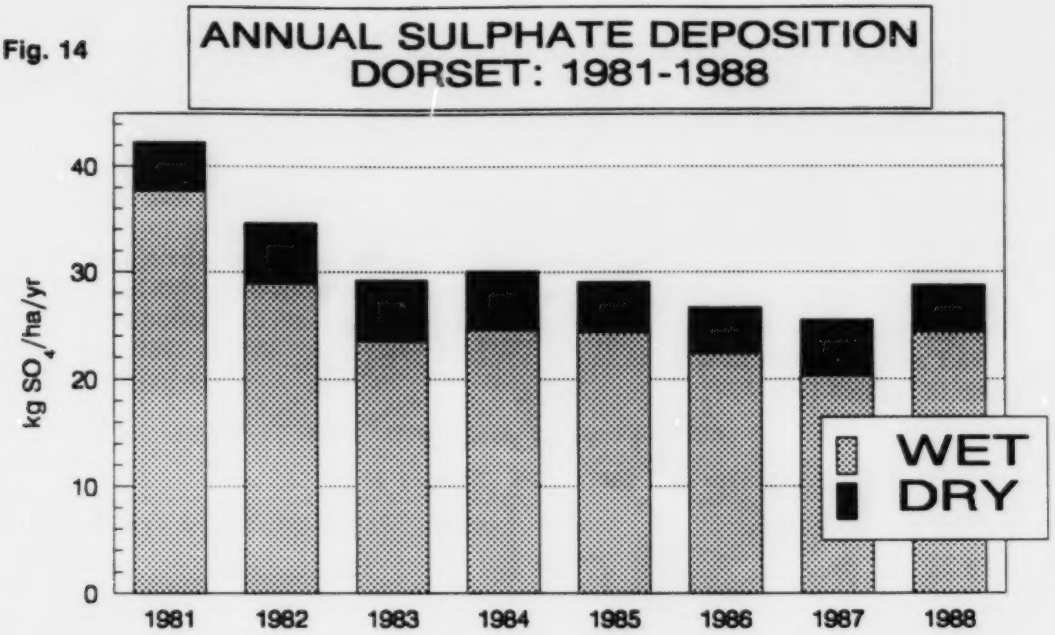
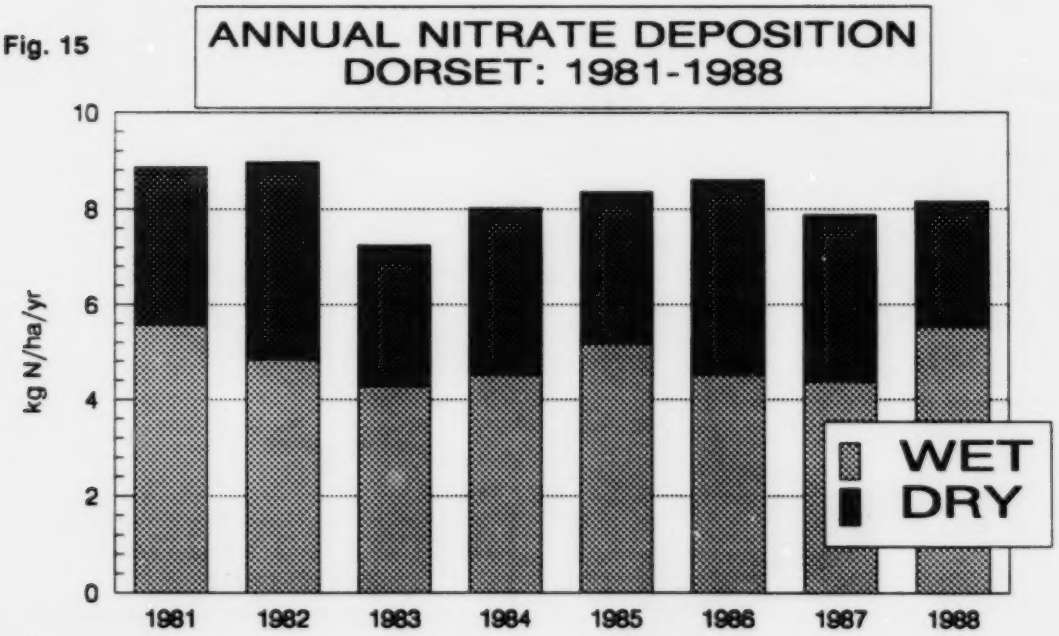

ANNUAL SULPHATE DEPOSITION
DORSET: 1981-1988
kg SO_4/ha/yr
0
10
20
30
40
1981
1982
1983
1984
1985
1986
1987
1988
WET
DRY

Fig. 15


ANNUAL NITRATE DEPOSITION
DORSET: 1981-1988
kg N/ha/yr
0
2
4
6
8
10
1981
1982
1983
1984
1985
1986
1987
1988
WET
DRY

9. ATMOSPHERIC DEPOSITION OF OTHER TOXIC SUBSTANCES - LEAD AND CADMIUM

As was mentioned in Section 2, the APIOS cumulative network also monitors the levels of various toxic trace metals. Among them, lead and cadmium are of particular current relevance, especially with respect to their atmospheric deposition to the Great Lakes basin. Deposition from the atmosphere is thought to be the major source of these metals in water from the Great Lakes (Strachan and Eisenreich, 1986). The annual average wet and dry deposition rates of lead and cadmium in Ontario (averaged over the period 1981 to 1988) are given in Figures 16 to 19.

Fig. 16

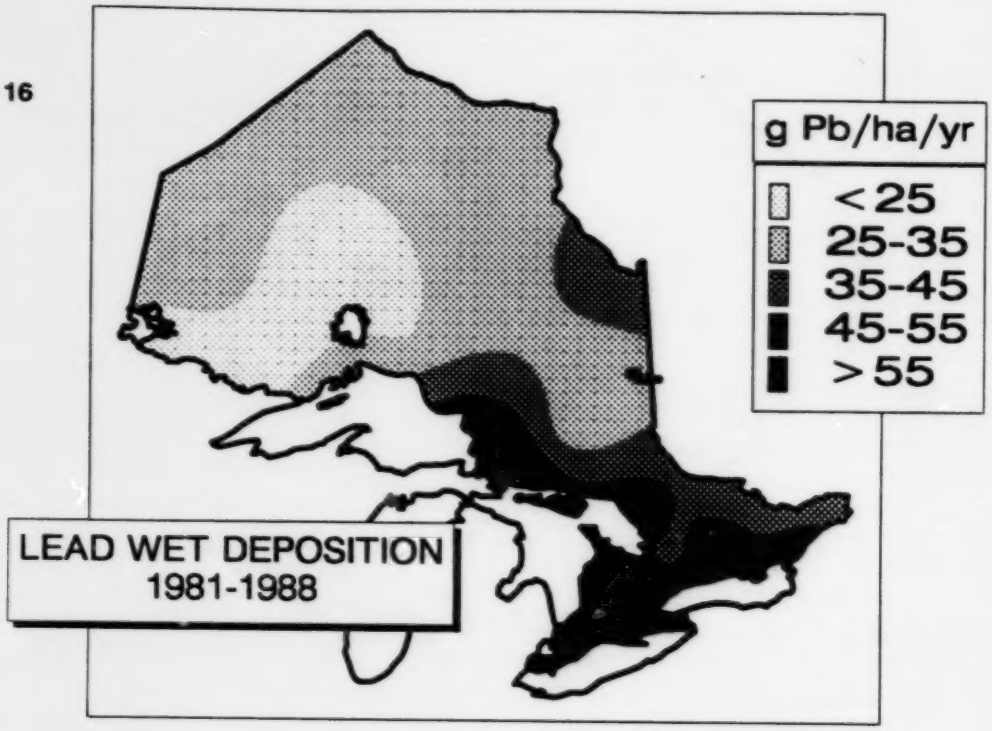

g Pb/ha/yr
< 25
25-35
35-45
45-55
> 55
LEAD WET DEPOSITION
1981-1988

Fig. 17

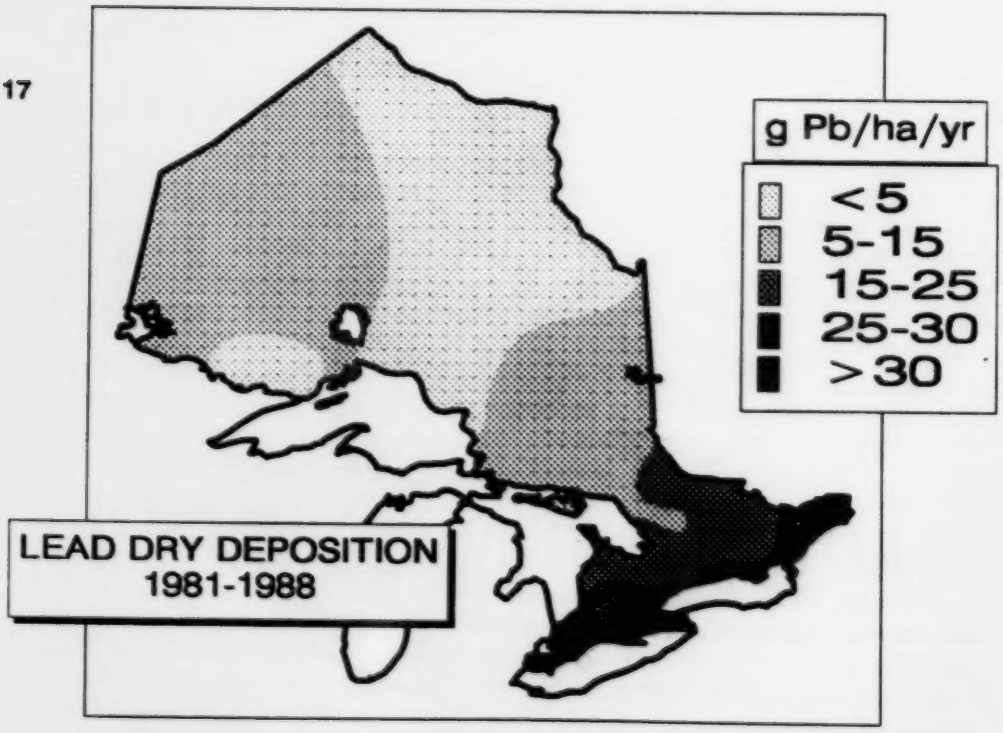

g Pb/ha/yr
< 5
5-15
15-25
25-30
> 30
LEAD DRY DEPOSITION
1981-1988

Fig. 18

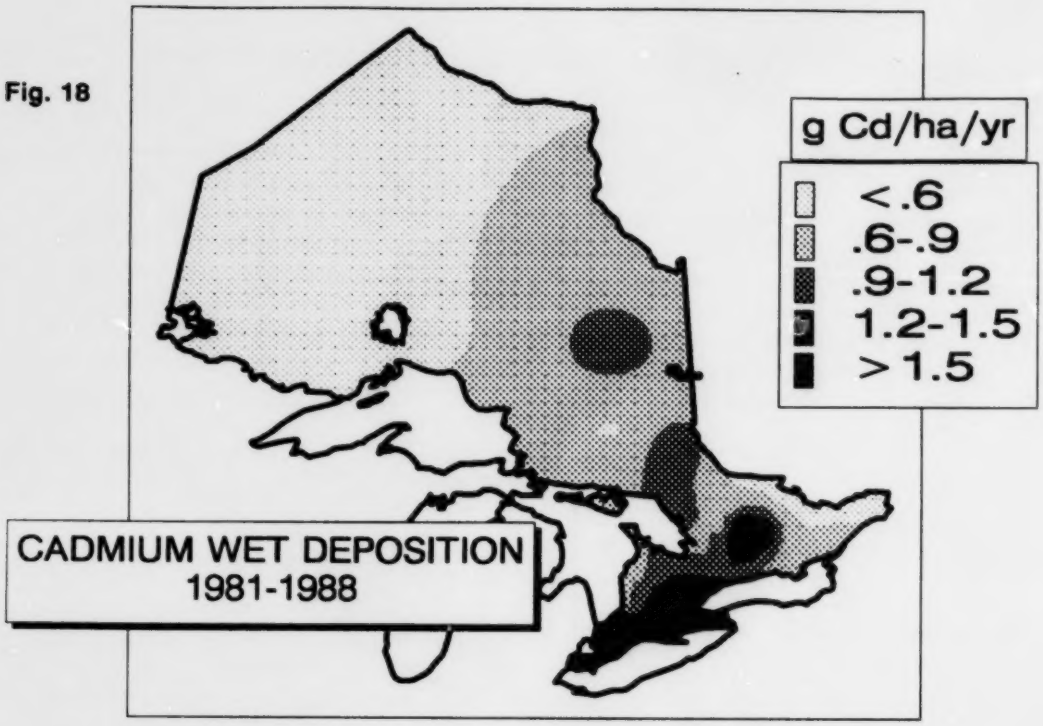
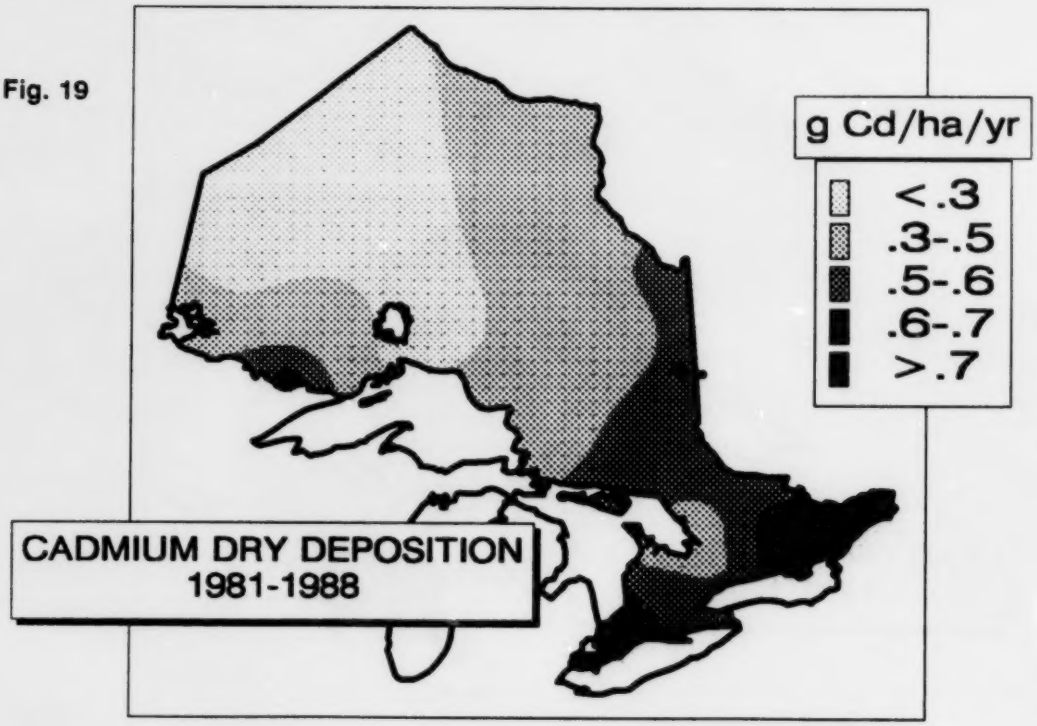

g Cd/ha/yr
< .6
.6-.9
.9-1.2
1.2-1.5
> 1.5
CADMIUM WET DEPOSITION
1981-1988

Fig. 19


g Cd/ha/yr
< .3
.3-.5
.5-.6
.6-.7
> .7
CADMIUM DRY DEPOSITION
1981-1988

10. CONTRIBUTION OF DIFFERENT EMISSION SOURCE AREAS TO ATMOSPHERIC DEPOSITION IN ONTARIO

The question, "How much does an emission source area (e.g. the states south of the Great Lakes) contribute to a particular receptor area (e.g. Algonquin Park)?" is of considerable interest to the acid rain problem. We now know that the sulphur and nitrogen oxides, which cause acidic deposition, can travel for hundreds of kilometres from their points of emission before being deposited on the earth's surface through wet and dry processes. Control of the acid rain problem will thus require both regional and local emission reductions. Ontario has already committed itself to an approximately 50% reduction of sulphur dioxide emissions by 1994 (compared to 1980 as the base year) and control of SO_2 emissions is included in the U.S. Clean Air Act Amendments of 1990. What effect is this reduction expected to have on acid rain in Ontario, due to the control programs being implemented in Canada and the United States?

One way to answer this question is by using mathematical models, which simulate the long-range transport, and atmospheric chemical transformation and deposition, of sulphur and nitrogen oxides. However, because of the complexity of atmospheric processes, many scientists feel that simple long-range transport models incorporating linear formulations of chemistry and deposition do not represent these processes sufficiently. To give an accurate answer, more detailed models (called long-range transport Eulerian models) are presently being evaluated. In the meantime, some useful answers can be obtained from a careful meteorological analysis of the monitoring data, especially data collected on an event-by-event basis.

Several such analyses have been carried out by the Ministry's scientists. The first example (Warmenhoven et al. 1991) assessed the contribution of Canadian and United States sources at monitoring sites near the border, using air parcel trajectory analyses. By this technique, the geographical areas over which the air parcel has passed during the previous day or two can be determined, and thus the portion of the measured atmospheric deposition due to emissions from the sources within different compass sectors can be assessed. Figures 20 and 21 show the results of such analysis at Longwoods and Charleston Lake, giving the percentage of the wet deposition of sulphates and nitrates due to USA, and Canadian sources. Although the results from border stations are particularly easy to interpret, since there is a clear-cut distinction between USA and Canadian source sectors, some air parcel trajectories (about 30% on average) still traverse both U.S. and Canadian areas during two days of travel, and are placed in the "unassignable" category. Nevertheless, Figures 20 and 21 indicate that at least 71% of the sulphate and nitrate wet deposition in southwestern Ontario, and at least 47% in southeastern Ontario, is due to emission in the United States. This result is not surprising. Southwestern Ontario, for example, forms a wedge extending into a heavily industrialized area of the United States. Within two days of air parcel travel time of Longwoods, the United States sector contains emission sources totalling about 16 million tonnes of SO_2 per year: the Canadian sector

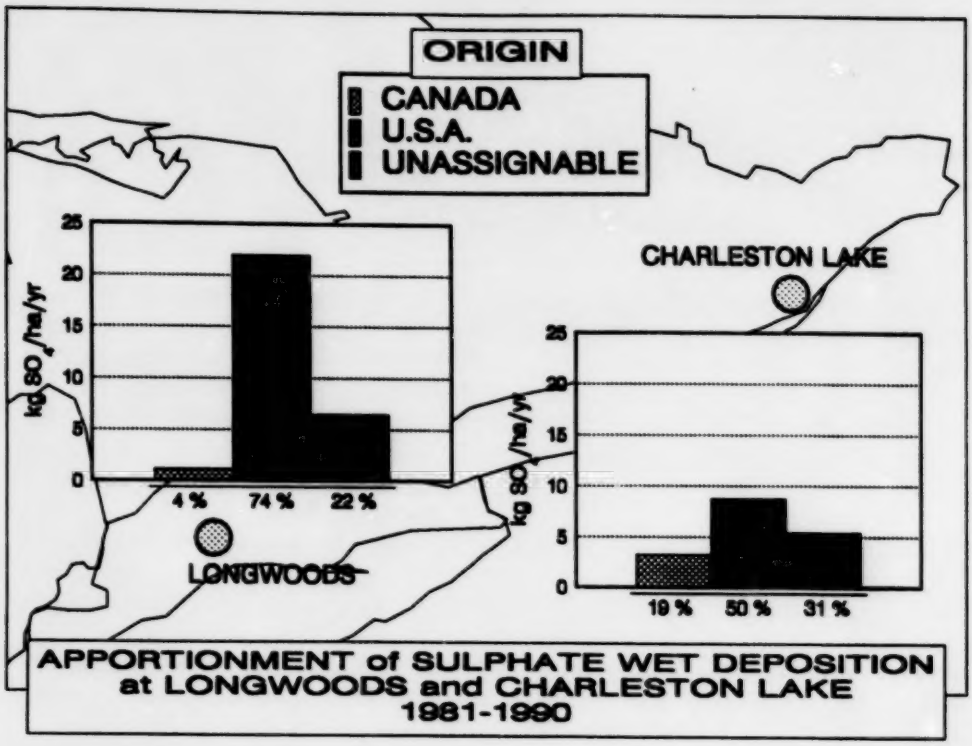
contains sources totalling only about 1.5 million tonnes. Thus the results of our source apportionment analysis are consistent with the emission information, and our knowledge of the residence time of sulphur and nitrogen oxides in the atmosphere. It should be noted that by the above analysis, we cannot distinguish between the atmospheric deposition due to anthropogenic and natural sources. If the contribution due to natural sources is subtracted away from the anthropogenic contribution, then the percentages in Figures 20 and 21 may change somewhat. However, it is known that anthropogenic emissions of sulphur and nitrogen oxides in northeastern North America considerably exceed natural emissions, so the change will not be large.

The results of another analysis tell a similar story. This analysis considered the impact of two large sources in Ontario - the INCO and Falconbridge smelters at Sudbury - on wet and dry deposition at various receptor areas. Together, these sources contributed about one-half of the total annual sulphur dioxide emissions in the province in 1980. However, during a prolonged shutdown of the smelters in 1982-1983, it was difficult to detect any decrease in the atmospheric deposition of sulphur oxides. A variety of meteorological analysis techniques were used. Figure 22 shows the results. The numbers in the circles are the estimated smelter contributions (% of annual total) of sulphate wet deposition at various receptor sites. The contribution was too small to be accurately quantifiable: they are expected to be "less than" the values shown in the circles. Most of the sulphur dry deposition comes from sulphur dioxide dry deposition, determined by two different types of analyses. One, a simple trajectory analysis, allowed direct estimation of the smelter contribution: the other, involving a more detailed meteorological analysis, gave upper limits only (indicated by the "less-than" values).

The implications of the above results are clear: the largest sulphur dioxide emission area in the province, Sudbury, contributes a relatively small portion of the total atmospheric deposition of sulphur oxides (less than 14% of the wet, and about 40% or less for dry deposition). The control of Ontario sources alone will not suffice. Without substantial concurrent emission reductions in the United States, acid deposition over large portions of the province will continue at unacceptable levels.

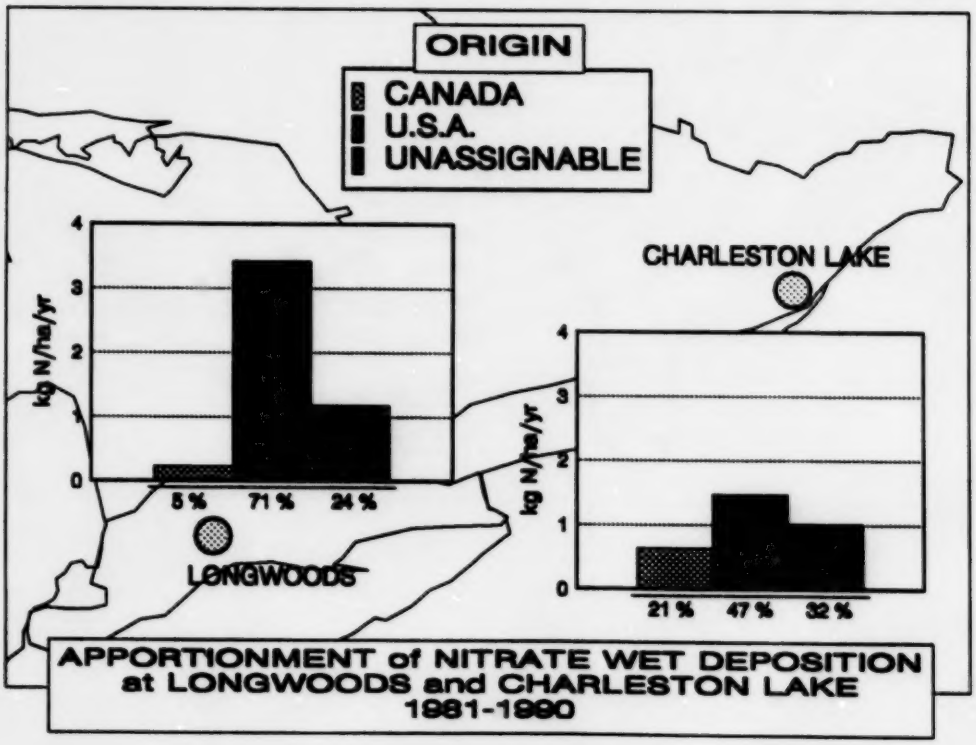
With both the Canadian and Unites States acid rain control programs now being implemented, it is expected that significant reductions in acid deposition will occur in sensitive receptor areas of Ontario, such as Muskoka.

Fig. 20



APPORTIONMENT of SULPHATE WET DEPOSITION
at LONGWOODS and CHARLESTON LAKE
1981-1990

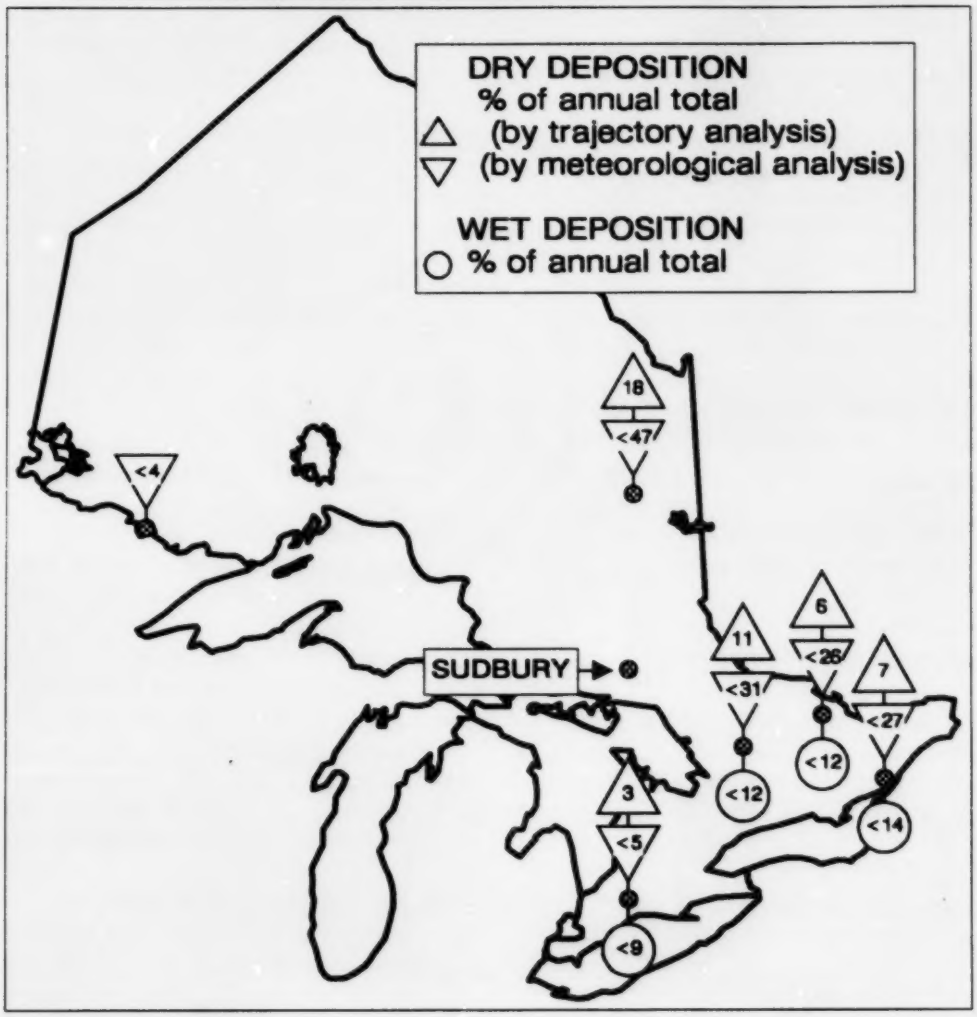
Fig. 21



APPORTIONMENT of NITRATE WET DEPOSITION
at LONGWOODS and CHARLESTON LAKE
1981-1990

Fig. 22

CONTRIBUTION of SUDBURY SMELTERS to the WET and DRY DEPOSITION of SULPHUR OXIDES in ONTARIO



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